

# Effect of Spin Correlations on Multi-orbital Metal-Insulator Transitions and Suppression of Orbital Selective Mott Transitions

Ya-Min Quan<sup>1</sup>, Liang-Jian Zou<sup>1,2\*</sup> and Hai-Qing Lin<sup>2</sup>

<sup>1</sup> Key Laboratory of Materials Physics,  
Institute of Solid State Physics, Chinese Academy of Sciences,  
P. O. Box 1129, Hefei 230031, China

<sup>2</sup> Department of Physics, Chinese University of Hong Kong,  
Shatin, New Territory, Hong Kong, China

(Dated: Nov 6, 2010)

We present the influence of spin correlation on the metal-insulator transitions (MIT) in two-orbital Hubbard models by the Kotliar-Ruckenstein slave-boson approach. In the asymmetric half-filling situation, the two orbits simultaneously transit from conducting to insulating states with the increase of Coulomb correlation, accompanied by a paramagnetic (PM)-antiferromagnetic (AFM) transition. The orbital selective Mott transition found in the PM condition is completely suppressed over a wide correlation range, though it may exist in the systems away from half-filling. In the insulating state, the system crosses over from a partially-polarized spin-gapped phase in the intermediate correlation regime to an almost fully-polarized Mott insulating phase in the strong correlation regime. These results demonstrate that the spin modulation to the quasiparticle spectra brings much rich and more interesting MIT scenario in multi-orbital correlated systems.

PACS numbers: 71.30.+h, 75.30.Kz, 71.10.Hf

Orbital selective Mott transition (OSMT), *i.e.*, with the increase of Coulomb correlation, a two-orbital electronic system transits from metal to a particular phase with one orbit exhibiting metallic conduction and another orbit being insulating, is an interesting topic<sup>1</sup>. Theoretically it has been known that such an unusual phase is robust in asymmetric two-orbital Hubbard models<sup>2-4</sup>. The OSMT phase can be driven by the asymmetric factors of two orbits, for example, the level splitting between two orbits and the different bandwidths or band degeneration. There exist great debates on whether some typical compounds with metal-insulator transitions (MIT), such as  $\text{Sr}_{2-x}\text{Ca}_x\text{RuO}_4$  and  $\text{V}_{2-x}\text{Cr}_x\text{O}_3$ , are the prototypes of the OSMT phase<sup>5-7</sup>. This naturally arises a question: whether does the OSMT phase really exist in realistic compounds, especially in the magnetic ordered systems? Generally speaking, the Mott-Hubbard MIT are accompanied with the change of magnetic structures. Such a change would crucially modulate the quasiparticle states, hence significantly affect the MIT. The MIT and OSMT phase have been studied widely based on the dynamical mean-field theory (DMFT)<sup>8</sup> and Kotliar-Ruckenstein slave boson (KRSB) methods<sup>9</sup>, both of which are effective many-body approaches to treat the electronic interaction over a wide correlation strength. However, limited by the huge computation time in the DMFT and KRSB methods, most of the studies focus on the paramagnetic (PM) phases. How the spin degree of freedom affects the MIT phase diagram and whether the OSMT phase exists in magnetic phase diagram are still not clear.

Hesagawa<sup>10</sup> and Sigrist *et al.*<sup>11</sup> first extended the single-orbital KRSB theory to the two-orbital situation to study the PM Mott transitions in two-orbital correlated electron sys-

tems. To explore the influence of the spin degree of freedom on the Mott transitions in multi-orbital Hubbard models, we apply the extended KRSB method<sup>8</sup> on asymmetric two-orbital Hubbard models for various electron fillings. With the help of a new numerical ansatz to the multi-orbital KRSB solution we developed recently<sup>12</sup>, which overcomes the convergency problem of many parameters in minimizing the groundstate energy, we could treat arbitrary interaction strengths, finite interorbital hoppings or hybridizations, and various magnetic configurations in multi-orbital correlated electron systems. In this Letter, starting with an asymmetric two-orbital Hubbard model, we present various magnetic phase diagrams of the MIT, and discuss the physical scenario of the MIT in the presence of spin degree of freedom. It is interestingly found that the OSMT phase completely vanishes at half filling. A Slater-type spin-gapped insulator and an orbital polarized band insulator are also found in magnetic phase diagrams, in addition to the conventional PM and *Néel* antiferromagnetic (AFM) metallic phases. Away from half filling, an AFM OSMT phase may be stable. These results show that the magnetic MIT picture in multi-orbital correlated systems is more interesting and much richer than that without spin.

Our starting point is an asymmetric two-orbital Hubbard model<sup>4,10</sup> applicable for various correlated electron systems

$$\begin{aligned}
 H &= H_0 + H_I \\
 H_0 &= - \sum_{i,j\alpha\beta\sigma} (t_{\alpha\beta} c_{i\alpha\sigma}^\dagger c_{j\beta\sigma} + h.c.) + \sum_{i\alpha\sigma} (\epsilon_\alpha - \mu) n_{i\alpha\sigma} \\
 H_I &= U \sum_{i\alpha} n_{i\alpha\uparrow} n_{i\alpha\downarrow} + \sum_{i\sigma\sigma'}^{(\alpha>\beta)} (U' - J_H \delta_{\sigma\sigma'}) n_{i\alpha\sigma} n_{i\beta\sigma'} \\
 &\quad - J_H \sum_{i\alpha\neq\beta} (c_{i\alpha\uparrow}^\dagger c_{i\alpha\downarrow} c_{i\beta\downarrow}^\dagger c_{i\beta\uparrow} - c_{i\alpha\uparrow}^\dagger c_{i\alpha\downarrow}^\dagger c_{i\beta\downarrow} c_{i\beta\uparrow}), \quad (3)
 \end{aligned}$$

on a square lattice with only the nearest-neighbor hopping integrals. Where  $t_{\alpha\beta}$ ,  $U$  ( $U'$ ) and  $J_H$  represent the hopping inte-

\*Correspondence author, Electronic mail: zou@theory.issp.ac.cn

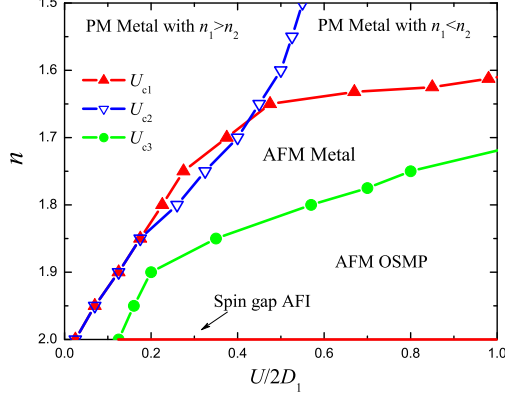


FIG. 1: Magnetic phase diagram of the two-orbital Hubbard model in the  $n-U$  plane.  $U_{c1}$  and  $U_{c3}$  denote the PM metal-AFM metal and the AFM metal-AFM insulator boundaries.  $U_{c2}$  separates different orbital polarization regions. Theoretical parameters:  $D_2/D_1 = 0.5$ ,  $J_H = 0.2U$  and  $\Delta = 0$ .

gral for the orbits  $\alpha$  and  $\beta$ , the intraband (inter-band) Coulomb repulsion and Hund's rule coupling, respectively. Throughout this paper we set  $U' = U - 2J_H$ , and take the energy bandwidth of the first orbit,  $2D_1 = 2\tau t_{11} = 2\tau t$ , be the energy unit. To explore the effect of spin in the multiorbital MIT, we introduce a few of auxiliary boson field operators representing the possibility of various electron occupations, such as  $e$ ,  $p_{\alpha\sigma}$ ,  $d_{\sigma\alpha\sigma'}$ ,  $b_{\alpha\beta}$ ,  $t_{\alpha\sigma}$  and  $q$ , which denote various possibilities of none, single, double, triplicate and quadruplicate electron occupations. Projecting the original fermion operators into these boson field and fermion field operators, one can obtain an effective Hamiltonian, from which one can get the groundstate energy in the saddle point approximation. We apply the generalized Lagrange multiplier method to enforce the fermion number constraint condition<sup>10</sup>. In this framework the interorbital hoppings and crystal field splitting can be treated in the same foot.

We adopt an optimizing method to get the minimized groundstate energy for four different magnetic configurations, including the PM or nonmagnetic, ferromagnetic (FM) and Néel AFM states. In the optimizing process the boson field normalization set a boundary for the mean value of each boson field. As shown in our previous work<sup>13</sup>, the optimizing problem of boundary constrained condition is still difficult. In numerically searching the global minima of the groundstate energy, we combine the pattern search method, the gradient method and the Rosenbrock method. As the optimizing point is approaching to the boundary, one must move one step inward the high-dimensional ellipsoid and the equipotential plane. Since the first axis of the new local orthogonal coordinate system of Rosenbrock method direct to the negative gradient direction, such an algorithm can be easily achieved. Our ansatz obtains a lot of desirable results for various PM systems with single, two and three orbitals<sup>12</sup>.

Fig.1 is magnetic phase diagrams of the asymmetric two-

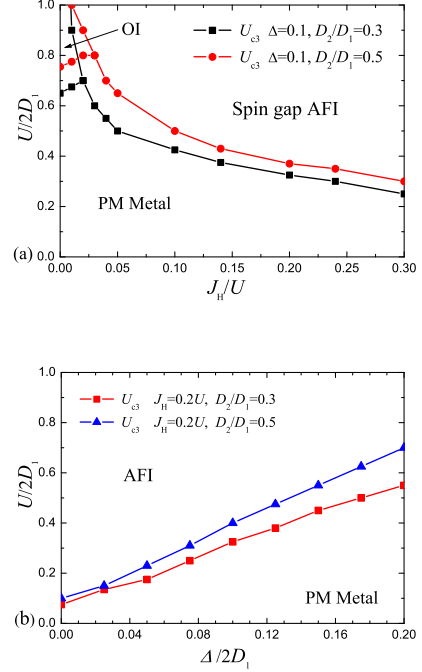


FIG. 2: Magnetic phase diagram of half-filled two-orbital Hubbard model in the  $U-J_H$  plane with  $\Delta/2D_1 = 0.1$  (a) and in the  $U-\Delta$  plane with  $J_H = 0.2U$  (b) for the bandwidth ratio  $D_2/D_1 = 0.3$  and  $0.5$ . OI and AFI denote the orbital insulator and the AFM insulator, respectively.

orbital Hubbard model with the bandwidth ratio  $D_2/D_1 = 0.5$ , the Hund's rule coupling  $J_H = 0.2U$  and the crystal field splitting  $\Delta = \varepsilon_2 - \varepsilon_1 = 0$ . We only plot the phase boundaries for the electron filling of  $n \leq 2$ , the magnetic phase diagram for  $n > 2$  can be mapped by considering the particle-hole symmetry character in the present square lattice with the nearest-neighbor hopping. On the whole, we find four different stable magnetic phases in Fig.1, including a PM metal, an AFM metal, an AFM OSMT phase and an AFM insulator. The boundaries between these different phases are  $U_{c1}$  and  $U_{c3}$ . Out of our expectation, the PM OSMT phase is not found for all of the electron filling and correlation strength. We address this magnetic phase diagram in length in the following.

Firstly, we find that in the half filling ( $n=2$ ) and  $\Delta=0$ , with the increase of the electron correlation the system undergoes transitions from a PM metal (only for  $U=0$ ) to an AFM metal at the the PM metal-AFM metal transition boundary  $U_{c1}$ , and then to an AFM insulator at the AFM MIT boundary  $U_{c3}$ , as seen in Fig.1. It is obviously that the critical line  $U_{c3}$  is considerable smaller than that without the spin degree of freedom<sup>14</sup>. Contrary to the early and recent results without the spin correlation<sup>14?</sup>, the OSMT phase is completely suppressed in the present situation. Since the spin degree of freedom is taken into account, the present AFM insulator is a spin gapped state in the intermediate correlation region, or a AFM Mott state in the large  $U$  region. Approximately, the spin gapped insulating region at half filling covers the PM OSMT

phase region obtained by the DMFT approach<sup>14</sup>. This shows that when  $U$  increases, the narrow orbit of the system becomes of AFM insulating. Such an AFM correlation also modifies the electronic properties in the wide band, and leads the spectra of the wide-band electron to open a gap. Hence the MIT simultaneously occurs in the two-orbital system. As a result, the PM OSMT phase is completely suppressed at half filling.

Away from half filling, the magnetic phase diagram becomes of more interesting. As seen in Fig.1, though the PM OSMT is completely suppressed, the AFM OSMT phase can survive over a wide region, in which the half-filled narrow band becomes of AFM and insulating, while the wide band away from half filling remains conducting, though modulated by the AFM correlation. We find that with the increase of the Coulomb correlation, the system undergoes a PM metal-AFM metal transition and an AFM metal-AFM OSMT transition. These transitions are also separated by the critical line  $U_{c1}$  and  $U_{c3}$ , respectively. Even in the PM phase, the system may show different orbital polarization separated by the critical line  $U_{c2}$ , as indicated in Fig.1. When  $U < U_{c2}$ , the particles are mainly distributed in the wide band and the reverse particle distributions achieve when  $U > U_{c2}$ , since the Coulomb interaction drives a charge redistribution with increasing  $U$ <sup>11,14</sup>. We notice that the critical line  $U_{c2}$  in Fig. 1 is almost independent of the magnetic transition, suggesting that the charge redistribution in different orbits is directly driven by the electron correlation, rather than the spin exchange splitting.

Though the PM OSMP is suppressed in the presence of magnetic correlation at half filling and  $\Delta = 0$ , it is unknown whether it exists over wide crystal field splitting  $\Delta$  and the Hund's rule coupling. In what follows, we focus on these situations in the half-filled Hubbard model. The magnetic phase diagrams for the half-filled asymmetric Hubbard models in the  $U - J_H$  and  $U - \Delta$  planes are plotted in Fig.2. It is still find that the OSMT phase is absent in Fig.2a and Fig.2b. Also, as seen in these two figures, the AFM metallic phase is unstable for finite crystal field splitting, it will become of a PM metallic one. Thus increasing the crystal field splitting may drive a AFM-PM transition.

Fig.2a shows the  $U - J_H$  phase diagram for different bandwidth ratios. In the PM metallic situation below the phase boundary  $U_{c3}$ , the electrons in two orbits are itinerant and equivalently occupy two orbits. The two orbits are singly occupied by two electrons and become insulating and the PM-AFM MIT occurs as  $U > U_{c3}$ . In the presence of spin degree of freedom, the system enters the spin-gapped AFM insulating phase in the intermediate region in Fig.2a. Due to the spin modulation, the PM-AFM MIT boundary  $U_{c3}$  is considerable smaller than that without spin. It is interesting that when the two orbits are split by a crystal field, a new insulating phase, named orbital insulator, appears in the small  $J_H$  region. In the orbital insulating phase, the two electrons fully occupy the lower narrow orbits and the system is nonmagnetic and enters into an insulating phase. Such an interesting phase has also been obtained recently by Yu and Si<sup>15</sup>. Due to the neglect of the spin effect in Yu and Si work, their  $U_c/2D_1$  is larger than unity, considerably larger than our result, about 0.68, in the system with  $J_H = 0$ ,  $D_2/D_1 = 0.3$  and  $\Delta = 0.1$ . With the

further increase of the electronic correlation, one may expect that the system crosses over from a spin-gapped insulator to a conventional Mott insulator, as we will show in Fig.3 later. These unusual results show that the spin correlation has profound influence on the quantum phases and the boarder of the MIT in correlated electron systems.

On the other hand, the magnetic phase diagram as the function of  $U$  and  $\Delta$  is shown in Fig.2b. One can see that the OSMT phase is also excluded. From the  $U$ - $\Delta$  phase diagram in Fig.2b, we see that the critical line  $U_{c3}$  of the PM-AFM MIT decreases with the decrease of the bandwidth ratios  $D_2/D_1$ . At the half filling, large Coulomb correlation and Hund's rule coupling, which contribute the spin exchange splitting, are needed to overcome the energy increase arising from the crystal field splitting. So the MIT boundary  $U_{c3}$  lifts with the increase of  $\Delta$ . It is interesting that the spin-gapped AFM insulator in Fig.1 and Fig.2 is not observed in the previous literature. In this phase the two orbits are not fully spin polarized, this allows spins to form density wave order. Considering the fact that this insulator is stable in the intermediate correlated regime, we attribute it to the *Slater*-type insulator<sup>16</sup> with a partial spin polarization.

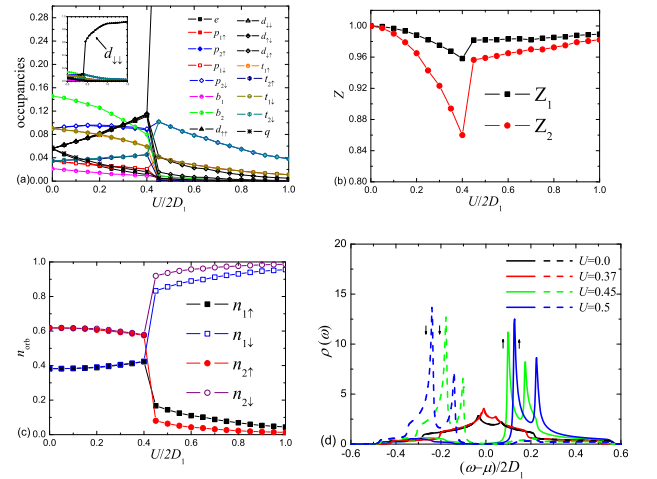


FIG. 3: Correlation dependence of occupation probabilities (a), band renormalization factor (b), particle number (c) and density of states (DOS) (d) of the half-filling two-orbital Hubbard model with  $D_2/D_1 = 0.5$ ,  $J_H = 0.2U$  and  $\Delta/2D_1 = 0.1$ . Inset in (a) displays the full data of the boson occupation probability.

Fig.3 shows the correlation dependence of occupation probabilities, band renormalization factor, particle number and DOS for finite Hund's rule coupling in the half-filled two-orbital Hubbard model. All these figures clearly show that the MIT occurs at the critical point  $U_{c3}$ , accompanied by a magnetic transition. Fig.3a displays the  $U$  dependence of various occupation probabilities. It shows that a first-order-type MIT transition occurs at  $U_{c3} = 0.42$ : when  $U < U_{c3}$ , various charge and spin configurations appear in approximately equal weight, suggesting that the PM metallic ground state is stable. When  $U > U_{c3}$ , the spin-triplet  $d_{\sigma\sigma}$  occupation is dominant, the system enters the magnetically ordered insulating phase.

As seen from Fig.3a, only the  $d_{\downarrow\downarrow}$  occupancy with each orbit occupied by one electron increases with the lift of  $U$  and  $J_H$ , the other occupancies decrease immediately when  $U > U_{c3}$ . After  $U \geq 2D_1$ , all of the occupancy probabilities almost approach constants. This indicates that the system enters a conventional Mott insulating phase.

Fig.3b shows the dependence of the band renormalization factor  $Z_\alpha$  of the quasiparticles in orbit- $\alpha$  on the Coulomb correlation  $U$ , which significantly differs from the behavior of  $Z$  without considering the spin correlation effect. In accordance with Fig.3a, we find that with the increase of  $U$ , the renormalization factor decreases in the PM phase since the electronic correlation narrows the bandwidths. Accompanied with the transition from the PM metallic phase to the AFM insulating one, an insulating spin gap appears. Out of our expectation, it abruptly increases at the MIT critical point  $U_{c3}/2D_1 = 0.42$ , and then gradually increases abnormally. With the increase of  $U/2D_1$  to about 1.0, both  $Z_1$  and  $Z_2$  approach saturations. The system may cross over from a Slater-type insulator to a conventional Mott-Heisenberg insulator, though both of the band renormalization factors do not show such a crossover. Our results are distinctly different from the PM results by Hasegawa<sup>10</sup> and Sigrist *et al.*<sup>11</sup>, where there exists a stable and robust OSMT phase in the intermediate correlation regime.

The  $U$  dependence of the particle number of each orbit per spin channel is shown in Fig.3c. It is clearly seen that with the increase of the electronic correlation, the particle numbers in the two orbits differ from each other in the PM region and experience a critical change at  $U_{c3}$ , and gradually approach constant occupations as  $U/2D_1$  approaches unity, in accordance with Fig.3a and Fig.3b. Obviously the PM-AFM MIT at  $U_{c3}$  is first order. Although the spins are not polarized in the PM metallic phase, the sublattice spins are strongly polarized in the insulating phase ( $U > U_{c3}$ ) and the sublattice spin polarization almost saturates as  $U/2D_1 \rightarrow 1$ . Asymmetric bandwidths and crystal field splitting lead the spin polarizations of two orbits be different. The more interesting is that with the transition from the PM to AFM phases when  $U > U_{c3}$ , the increase of the Coulomb interaction depresses the orbital polarization, which originates from the increase of the Hund's rule coupling. We find that the finite crystal field splitting is the key reason for the first-order MIT. When the crystal field splitting equals to zero, the transition from the PM to magnet-

ically ordered phases is the second order, in agreement with Hasegawa's results<sup>10</sup>.

The spin-dependent densities of states (DOS) for  $U/2D_1 = 0, 0.37, 0.45$  and  $0.5$  are plotted in Fig.3d. It demonstrates how the two-orbital DOS evolve with the electronic correlation. In the PM metallic phase, both bandwidths of the two orbits shrink with increasing  $U$ . In the present square lattice, one finds two cusps in the spin-dependent DOS of the two orbits when the crystal field splitting is switched on. The MIT energy gap, which becomes large with the increase of  $U$ , opens at  $U_{c3} = 0.42$  in the system with  $D_2/D_1=0.5$ ,  $J_H=0.2U$  and  $\Delta/2D_1=0.1$ . The well-known quasiparticle peaks in Fermi surface and the three-peaks structure found in the DMFT approach are not observed in the spin-dependent multi-orbital KRSB theory, which is attributed to the neglect of dynamical transition process between Hubbard subbands in the present mean-field approach.

From the preceding studies on the metal-insulator transitions and the orbital selective Mott phase in the two-orbital Hubbard model with spin degree of freedom in the slave boson mean field theory, we find that in the presence of spin correlation, the scenario of the metal-insulator transitions may be distinctly different from that in the paramagnetic situations. In the half-filled asymmetric two-orbital Hubbard models, the spin modulation arising from the insulating narrow band opens a gap in the wide band, leading the paramagnetic orbital selective Mott phase to be suppressed over a wide correlation range; and the orbital insulating phase is also found to stable in the small  $J_H$  region. In the presence of spin correlations and finite crystal field splitting, the metal-insulator transitions, accompanied by magnetic transitions, are of first order. Away from half-filling an antiferromagnetic orbital selective Mott phase may survive and the critical point of  $U_{c3}$  is smaller than the PM results because the symmetry of the local interaction is lowered by the spin polarization. These results demonstrate that the spin correlations and the crystal field splitting play very important roles for the metal-insulator transitions.

This work was supported by the NSFC of China, the HK-SAR RGC 401806 and the Knowledge Innovation Program of the Chinese Academy of Sciences. Numerical calculations were performed at the Center for Computational Science of CASHIPS.

<sup>1</sup> V. I. Anisimov, I.A. Nekrasov, D.E. Kondakov, T.M. Rice and M. Sigrist, *Euro. Phys. J. B* **25**, 191 (2002).

<sup>2</sup> A. Koga, N. Kawakami, T.M. Rice and M. Sigrist, *Phys. Rev. Lett.* **92**, 216402 (2004); A. Koga, N. Kawakami, T.M. Rice and M. Sigrist, *Phys. Rev. B* **72**, 045128 (2005); A. Koga, K. Inaba and N. Kawakami, *Prog. Theo. Phys. Suppl.* **160**, 253 (2005); K. Inaba and A. Koga, *Phys. Rev. B* **73**, 155106 (2006).

<sup>3</sup> J. Buenemann, D. Rasch and F. Gebhardet, *J. Phys. Cond. Matt.* **19**, 436206 (2007).

<sup>4</sup> Y. Song and Liang-Jian Zou, *Phys. Rev. B* **72**, 085114 (2005).

<sup>5</sup> M. Neupane, P. Richard, Z.-H. Pan, Y. Xu, R. Jin, D. Mandrus, X. Dai, Z. Fang, Z. Wang and H. Ding, *Phys. Rev. Lett.* **103** 097001

(2009).

<sup>6</sup> A. Liebsch and H. Ishida, *Phys. Rev. Lett.* **98**, 216403 (2007).

<sup>7</sup> Y. Song and Liang-Jian Zou, *Euro. Phys. J. B* **72**, 59 (2009).

<sup>8</sup> A. Georges, G. Kotliar, W. Krauth and M. J. Rosenberg, *Phys. Rev. Lett.* **57**, 1362 (1986).

<sup>9</sup> G. Kotliar and A. Ruckenstein, *Phys. Rev. Lett.* **57**, 1362 (1986).

<sup>10</sup> H. Hasegawa, *J. Phys. Soc. Jpn.* **66**, 1391 (1997); *Phys. Rev. B* **56**, 1196 (1997).

<sup>11</sup> A. Rüegg, M. Indergand, S. Pilgram and M. Sigrist, *Eur. Phys. J., B* **48** 55 (2004).

<sup>12</sup> Y. M. Quan, Dayong Liu and Liang-Jian Zou, to appear in *Acta Physica Sinica* 2011 (Chinese).

- <sup>13</sup> Y. M. Quan, Liang-Jian Zou, Dayong Liu and H. Q. Lin, arXiv:1104.4599.
- <sup>14</sup> E. Jakobi, Nils Blümer and Peter van Dongen, Phys. Rev. B **80**, 115109 (2009).
- <sup>15</sup> Rong Yu and Qimiao Si, arXiv:1006.2337v2.
- <sup>16</sup> F. Gebhard, *The Metal-Insulator Transition: Models and Methods*, Springer, (1997).